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OMB No 0704-0188

1. AGENCY USE ONLY (Leave blank)

DATE
13

3. REPORT TYPE AND DATES COVERED
Reprint

2

4. TITLE AND SUBTITLE
Displacive Excitation of Coherent Phonons

5. FUNDING NUMBERS
DAAL03-92-C-0001

6. AUTHOR(S)
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8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
U. S. Army Research Office
P. O. Box 12211
Research Triangle Park, NC 27709-2211

10. SPONSORING/MONITORING AGENCY REPORT NUMBER
ARO 28925.22-EL

11. SUPPLEMENTARY NOTES
The view, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy, or decision, unless so designated by other documentation.

12a. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution unlimited.

12b. DISTRIBUTION CODE

13. ABSTRACT (Maximum 200 words)

93-25060

14. SUBJECT TERMS

15. NUMBER OF PAGES

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
UNCLASSIFIED	UNCLASSIFIED	UNCLASSIFIED	UL

Displacive Excitation of Coherent Phonons

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Recently, we have shown that coherent lattice vibrations can be induced and subsequently detected in certain semimetals and semiconductors (e.g. Bi, Sb, Te, Ti_2O_3) via time-resolved optical pump-probe measurements using a CPM oscillator.[1] The experimental data show in each case that only totally symmetric lattice modes (i.e., A_1 symmetry) are coherently excited, even though other symmetry modes of comparable Raman cross-section exist. Furthermore, careful measurement of the coherent phonon phase reveals that the excitation mechanism for coherent phonons in these materials appears to be qualitatively different from that responsible for impulsive stimulated Raman scattering previously seen in organic dyes and crystals.

A phenomenological model has been constructed for coherent phonon generation in the materials under study and is referred to as displacive excitation of coherent phonons (DECP).[2] In the DECP model, we argue that the quasi-equilibrium coordinates of only the totally symmetric phonon modes are "instantaneously" shifted due to electronic excitation by the optical pump pulse. The lattice, finding itself displaced from its excited state quasi-equilibrium, undergoes coherent cosinusiodal oscillation about its new equilibrium position until the oscillation is damped out via electron-phonon and/or phonon-phonon scattering events. The fit of the experimental data to the DECP model is excellent.

Here, this technique for generating coherent phonons is used to investigate the temperature dependent behavior of Ti_2O_3. It is well-known that between 300K to 570K dramatic changes take place in Ti_2O_3 with respect to both lattice and electronic properties, turning it from a semiconductor to a metal. Temperature dependent X-ray measurements reveal a significant deformation of its corundum structure along the A_{1g} symmetry lattice coordinate, but with no change in lattice symmetry. Transport measurements indicate a smoothly varying drop in its resistivity by over a factor of 30. Spontaneous Raman scattering measurements show a softening of the optical phonon frequencies by up to 10%.

In the figure we show the temperature dependent pump-probe data for Ti_2O_3. The pronounced oscillatory components in the data are the signatures of the lower frequency Raman-active A_{1g} phonon mode. We observe that this mode frequency decreases (7.0 THz -> 6.2 THz) as a function of increasing temperature consistent with conventional Raman results.

Because the reflectivity changes are so large, it is of interest to estimate the magnitude of the actual phonon vibrational amplitude in Ti_2O_3. By simply assuming that there exists a mapping between static equilibrium properties and the time-resolved properties in Ti_2O_3, we can estimate the magnitude of the vibrational amplitude. This calibration would be provided by temperature dependent X-ray and optical reflectivity measurements. Temperature dependent X-ray data can accurately measure the anomalous changes in the Ti-Ti and Ti-O separations as a function of increasing temperaure. Rice and Robinson have found that through the metal-insulator transition, the ions move along a vibrational coordinate of A_{1g} symmetry, the same which we coherently excite.[3] Accompanying this change in lattice parameters is also a marked change in the optical reflectivity of the material which has been measured as a function of temperature. By relating the reflectivity and X-ray data through their temperature dependences, a mapping can be established between the static reflectivity of the material and the magnitude of the Ti-Ti separation. From 400K to 500K, we have measured in a separate steady-state experiment that a 10% decrease in the optical reflectivity at 2 eV corresponds to an increase in the nearest neighbor Ti-Ti separation by a distance of ~ 0.06Å. If the phonon dynamics of the system can be assumed to appear quasistatic to the electrons on the timescale of the A_{1g} oscillation period, then a 5% change in transient reflectivity due to the coherent phonon (obtained by deconvolving the raw data with the pulse autocorrelation) would correspond to a

Springer Series in Chemical Physics, Vol. 55 **Ultrafast Phenomena VIII**
Editors: J.-L. Martin · A. Migus · G.A. Mourou · A.H. Zewail

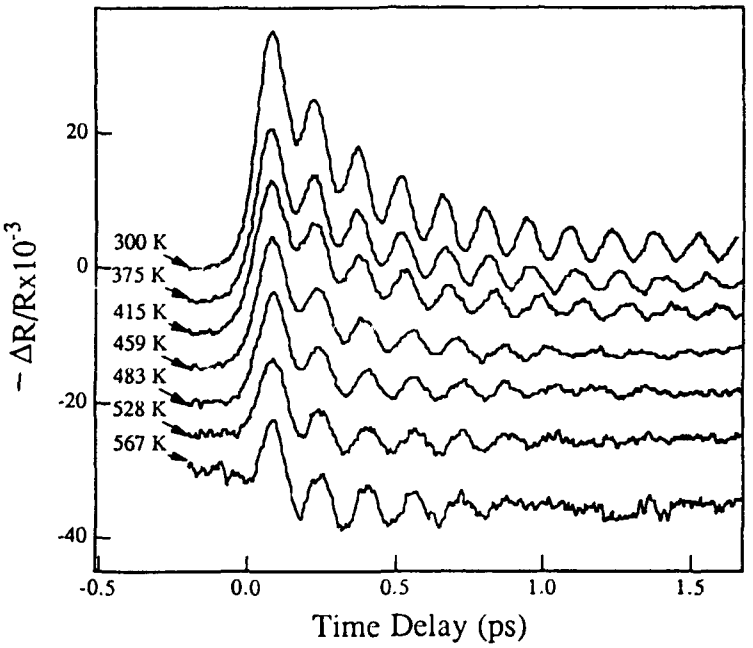

Figure : temperature consistent with conventional Raman results.

vibrational amplitude of 0.03 Å (1.2 % change in Ti-Ti separation). It must be noted that the accuracy of this mapping relies on the assumption that temperature dependent changes in refractive index in the equilibrium measurements arise primarily from deformations along A_{1g} phonon coordinates and not other phonon coordinates.

Vibrational amplitudes of this size suggest that modulating an insulator-metal transition at THz frequencies is possible. The band model parameters for semiconductors are dependent upon the lattice parameters of the crystal via the deformation potential. This can be verified experimentally by pressure dependent energy gap measurements. In a coherent phonon generation experiment, it is possible to induce a coherent lattice vibration of large amplitude as estimated above. We propose that this modulation of the lattice parameter can in turn modulate the energy bands of a material (and thus the energy gap). For an appropriate bias temperature (so that the energy gap is very small) and a coherent lattice vibration of sufficient amplitude, the conduction and valence bands in the material can be made to overlap at a frequency of ~ 7 THz. Whether or not transport properties are modulated at ~ 7 THz depends on the dynamical equilibration time for intervalley carrier scattering between the valence and conduction band. This simple picture, of course, assumes that the rigid band approximation is valid.

This work was supported in part at MIT by JSEP DAAL03-92-C-0001, AFOSR F49620-91-C-0091, and ONR 76804N0001492J1439. TKC ackowledges support of a doctoral fellowship by the IBM Corporation.

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